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<b>(54) Title:</b> THERMOPLASTIC TRANSPARENT RESIN COMPOSITION AND METHOD OF MANUFACTURING SAME <b>(57) Abstract</b> <p>Disclosed is a thermoplastic transparent resin composition and a method for manufacturing the same. The resin composition includes 3-15 parts by weight of small aperture polybutadiene rubber latex; 5-25 parts by weight of large aperture polybutadiene rubber latex; 40-70 parts by weight of a methacrylic acid alkylester compound or an acrylic acid alkylester compound; 15-30 parts by weight of an aromatic vinyl compound; and 1-20 parts by weight of a vinylcian compound. The method includes the steps of a) producing a small aperture polybutadiene rubber latex having an average particle diameter of 600-1500 Å, a gel content of 70-95 %, and a swelling index of 12-30 by reacting butadiene at 55-70 °C using a polymerization initiator; b) producing a large aperture polybutadiene rubber latex having a particle diameter of 2600-5000 Å, a gel content of 70-95 %, and a swelling index of 12-30 by enlarging the small aperture polybutadiene rubber latex particles; c) performing graft copolymerization at 65-80 °C by continuously or separately adding 3-15 parts by weight of the small aperture polybutadiene rubber latex of step a), 5-25 parts by weight of the large aperture polybutadiene rubber latex of step b), 40-70 parts by weight of a methacrylic acid alkylester compound or an acrylic acid alkylester compound, 15-30 parts by weight of an aromatic vinyl compound, and 1-20 parts by weight of a vinylcian compound.</p>		

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# THERMOPLASTIC TRANSPARENT RESIN COMPOSITION AND METHOD OF MANUFACTURING SAME

## BACKGROUND OF THE INVENTION

### (a) Field of the Invention

5           The present invention relates to a thermoplastic transparent resin composition, and more particularly, to a thermoplastic transparent resin composition which is resistant to shocks, chemically resistant, easily processed, and highly transparent. The present invention also relates to a method for manufacturing the thermoplastic transparent resin composition in which, when  
10 performing graft copolymerization of a methacrylic acid or an acrylic alkylester compound, an aromatic vinyl compound, and a vinylcyan compound in a mixture of a small aperture polybutadiene rubber latex and a large aperture polybutadiene rubber latex, both having a suitable gel content, a refraction coefficient is maximized by controlling the mixture ratio of these monomers,  
15 thereby realizing the advantages of the resin composition stated above.

### (b) Description of the Related Art

Many various products today use transparent plastic parts for both practical reasons and to provide distinguishable characteristics to a product. Examples include the plastic screen cover on a cell phone, a washing machine  
20 cover that enables the user to view the contents being washed, portions of the housing of a PC monitor, pager housings, the housings and covers of home appliances, etc. However, the typically used ABS (acrylonitrile-butadiene-styrene) copolymer resin, although providing good shock resistance, chemical

resistance, processability, and surface gloss, has limited transparency.

The following are some of the methods for providing transparency in plastic materials:

1) A method using transparent polycarbonate resin.

5        2) A method of providing shock resistance to transparent PMMA (polymethylmethacrylate) resin (U.S. Patent No. 3,787,522, Japanese Patent No. Sho 63-42940).

3) A method of providing transparency to HIPS (high impact polystyrene) resin (European Patent No. 0,703,252).

10        However, in the method of using polycarbonate resin, although this material has good transparency and shock resistance at room temperature, it is not highly resistant to chemicals and does not display high resistance to shocks at low temperatures. This material also has problems related to processability, making it difficult to manufacture this product to large sizes. With regard to  
15        PMMA resin, though providing good transparency and processability, this material has an extremely low level of resistance against shocks. Finally, regarding HIPS resin, this material has a low level of resistance against chemicals and is easily scratched.

U.S. Patent No. 4,767,833, in an effort to solve the above problems,  
20        discloses a transparent resin in which monomers such as methylemethacrylate, styrene and acrylonitrile are graft copolymerized in SBR (styrene-butadiene rubber) latex such that properties of shock resistance, chemical resistance, and processability are realized. However, this resin is limited in its resistance to

shocks at low temperatures and displays a limited degree of transparency.

### **SUMMARY OF THE INVENTION**

The present invention has been made in an effort to solve the above  
5 problems.

It is an object of the present invention to provide a thermoplastic transparent resin composition and a method of manufacturing the same in which the resin composition displays high resistance to shocks at both room temperature and low temperatures, high chemical resistance, good  
10 processability, high surface gloss, and excellent transparency.

To achieve the above object the present invention provides a thermoplastic transparent resin composition comprising 3-15 parts by weight of small aperture polybutadiene rubber latex, 5-25 parts by weight of large aperture polybutadiene rubber latex, 40-70 parts by weight of a methacrylic acid  
15 alkylester compound or an acrylic acid alkylester compound, 15-30 parts by weight of an aromatic vinyl compound, and 1-20 parts by weight of a vinylcian compound.

Further, the present invention provides a method of manufacturing a thermoplastic transparent resin composition including the steps of a) producing a  
20 small aperture polybutadiene rubber latex having an average particle diameter of 600-1500 Å, a gel content of 70-95%, and a swelling index of 12-30 by reacting butadiene at 55-70°C using a polymerization initiator, b) producing a large

aperture polybutadiene rubber latex having a particle diameter of 2600-5000 Å, a gel content of 70-95%, and a swelling index of 12-30 by enlarging the small aperture polybutadiene rubber latex particles, and c) performing graft copolymerization at 65-80°C by continuously or separately adding 3-15 parts by weight of the small aperture polybutadiene rubber latex of step a), 5-25 parts by weight of the large aperture polybutadiene rubber latex of step b), 40-70 parts by weight of a methacrylic acid alkylester compound or an acrylic acid alkylester compound, 15-30 parts by weight of an aromatic vinyl compound, and 1-20 parts by weight of a vinylcian compound.

10

#### **DETAILED DESCRIPTION OF THE PRESENT INVENTION**

In the present invention, when producing an ABS resin comprised of acrylonitrile having high chemical resistance, butadiene having good resistance to shocks, and styrene providing good processability, methylmethacrylate is introduced, and by controlling a content and mixture ratio of each component, a refraction coefficient of polybutadiene rubber latex and a refraction coefficient of a mixture of the methylmethacrylate, styrene, and acrylonitrile, grafted in the rubber latex, are maximized. Accordingly, a thermoplastic transparent resin is manufactured having high levels of shock resistance, chemical resistance, and processability, in addition to excellent transparency.

The thermoplastic transparent resin composition of the present invention is manufactured by graft copolymerizing, using an emulsion polymerization method, a mixture of a small aperture polybutadiene rubber having a particle

diameter of 600-1500 Å, a gel content of 70-95%, and a swelling index of 12-30 and a large aperture polybutadiene rubber latex having a particle diameter of 2600-5000 Å, a gel content of 70-95%, and a swelling index of 12-30, and monomers grafted in the mixture of a methacrylic acid or an acrylic acid alkylester compound, an aromatic vinyl compound, and a vinylcian compound.

The particle diameter and gel content of the polybutadiene rubber latex used in the present invention greatly influence properties of the inventive resin composition such as transparency and resistance to shocks. Therefore, particle diameter size and gel content must be suitably controlled to ensure that optimal levels of such properties are realized. That is, in the case of the rubber latex particle size, although transparency is increased with decreases in particle size, shock resistance and fluidity are decreased, whereas the opposite results with increases in particle size. Further, if the gel content of the rubber latex is low, since the monomers undergo significant swelling in the rubber latex during graft reaction, an external particle diameter of the rubber latex is increased. This limits transparency but improves resistance to shocks. Here also, the opposite effect results with increases in the gel content.

Further, a refractive coefficient of the monomer mixture is directly related to transparency of the inventive resin composition. The refractive coefficient is controlled by a mixture ratio of the monomers. That is, since a refractive coefficient of polybutadiene is approximately 1.518, the remaining grafted components must be similar to this level to realize a high level transparency. Refractive coefficients for each component are 1.49 for the methylmethacrylate,



1.59 for the styrene, and 1.518 for the acrylonitrile.

During the graft copolymerization reaction, each component can be added all at once, or each separately (or partially for each component) in a continuous manner. The present invention performs the adding of components using both these methods. This will be described in more detail hereinafter.

A method of manufacturing the thermoplastic transparent resin composition of the present invention will now be described.

#### 1) Manufacture of polybutadiene rubber latex

##### a) Manufacture of small aperture rubber latex

100 parts by weight of 1,3-butadiene, 1-4 parts by weight of a emulsifying agent, 0.1-0.6 parts by weight of a polymerization initiator, 0.1-1.0 parts by weight of an electrolyte, 0.1-0.5 parts by weight of a molecular weight controlling agent, and 90-130 parts by weight of ion exchange water are added all at once, and reacted at 50-65°C for 7-12 hours. Next, 0.5-1.2 parts by weight of a molecular weight controlling agent is further added and reacted at 55-70°C for 5-15 hours, thereby producing small aperture polybutadiene rubber latex having an average particle diameter of 600-1500 Å, a gel content of 70-95%, and a swelling index of 12-30.

As the emulsifying agent, it is possible to use one or a mixture of two or more of the following: alkyl aryl sulfonate, alkali methyl alkyl sulfate, sulfonated alkylester, fatty acid soap, and rosin acid alkali salt. As the polymerization initiator, it is possible to use a water soluble persulfate or peroxy compound, or an oxidation-reduction compound. The most suitable water soluble persulfates

are sodium and potassium. For a fat soluble polymerization initiator, it is possible to use cumene peroxide, diisopropyl benzene hydroperoxide, azobisisobutyronitrile, tertiary butyl hydroperoxide, paramethane hydroperoxide, and benzoyl peroxide. For the electrolyte, it is possible use one or a mixture of two or more of the following: KCl, NaCl, KHCO<sub>3</sub>, NaHCO<sub>3</sub>, K<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>CO<sub>3</sub>, KHSO<sub>3</sub>, NaHSO<sub>3</sub>, K<sub>4</sub>P<sub>2</sub>O<sub>7</sub>, K<sub>3</sub>PO<sub>4</sub>, Na<sub>3</sub>PO<sub>4</sub>, K<sub>2</sub>HPO<sub>4</sub>, and Na<sub>2</sub>HPO<sub>4</sub>.

An important factor in determining gel content and the swelling index of the rubber latex is polymerization temperature. The type of initiator used also influences these two characteristics of the rubber latex.

10           b) Manufacture of large aperture rubber latex (small aperture rubber latex adhesion process)

3.0-4.0 parts by weight of an acetic acid aqueous solution is slowly added over a period of one hour to 100 parts by weight of the small aperture rubber latex having a particle diameter of 600-1500 Å, a gel content of 70-95%, and a swelling index of 12-30, thereby enlarging the particle size. Stirring is then discontinued and a large aperture rubber latex results, the large aperture rubber latex having a particle diameter of 2600-5000 Å, a gel content of 70-95%, and a swelling index of 12-30.

To provide shock resistance to the rubber latex, although it is possible to manufacture the large aperture rubber latex using a direct polymerization method (Japanese Laid-Open Patent No. 56-136807), a long reaction time is required and the gel content level that can be obtained is limited with this method. Accordingly, in order to produce a large aperture rubber latex in a short

amount of time while maintaining a high gel content, it is preferable, as described above, to first manufacture a small aperture rubber latex having a high gel content, then add an acidic material to the rubber latex to enlarge the particles, thereby producing a large aperture rubber latex.

5 2) Manufacture of graft copolymerization material

Graft copolymerized in a mixture of 3-15 parts by weight of the small aperture polybutadiene rubber latex and 5-25 parts by weight of the large aperture polybutadiene rubber latex are 40-70 parts by weight of a methacrylic acid alkylester compound or an acrylic acid alkylester compound, 15-30 parts by weight of an aromatic vinyl compound, 1-20 parts by weight of a vinylcian  
10 compound, 0.2-0.6 parts by weight of an emulsifying agent, 0.2-0.6 parts by weight of a molecular weight controlling agent, and 0.05 to 0.3 parts by weight of a polymerization initiator.

In the above, a suitable polymerization temperature is 65-80°C, and a  
15 suitable polymerization time is 4-7 hours. In the polymerization reaction, it is possible to use methymethacrylate for the methacrylic acid alkylester compound or acrylic acid alkylester compound; for the aromatic vinyl compound, it is possible to use styrene,  $\alpha$ -methylstyrene, o-ethylstyrene, p-ethylstyrene, or vinyl toluene; and for the vinylcian compound, it is possible to use acrylonitrile,  
20 methacrylonitrile, or ethacrylonitrile.

As the emulsifying agent used in the polymerization reaction, it is possible to use one or a mixture of two or more of alkyl aryl sulfonate, alkali methyl alkyl sulfate, sulfonated alkylester, fatty acid soap, and rosin acid alkali

salt. For the molecular weight controlling agent, tertiary dodecyl mercaptan is generally used. As the polymerization initiator, it is possible to use an oxidation-reduction catalyst realized through a mixture of (a) a peroxide such as cumene peroxide, diisopropyl benzene hydroperoxide, and persulfuric acid salt, and (b) a  
5 reducing agent such as sodium formaldehyde sulfoxylate, sodium ethylenediaminetetraacetic, primary iron sulfate, dextrose, pyrroline acid sodium, and sulfurous sodium.

After polymerization is complete, a polymerization conversion rate of the obtained latex is over 98%. An oxidation preventing agent and a stabilizer is  
10 then added to the latex, and at a temperature over 80°C, the latex is coagulated using a calcium chloride aqueous solution, after which the latex is desiccated and dried, thereby obtaining a powder.

Stability of the graft copolymerized latex manufactured in the above is determined by measuring a solidification rate (%) using Equation 1 below.

15 [Equation 1]

Solidification(%) = produced solidification wt.(g) in reactor/ weight of all rubber and monomers X 100

A solidification rate of over 0.7% is indicative of an extremely low latex stability, and there will occur difficulty in realizing suitable graft copolymerization  
20 as a result of the large presence of solid material.

After adding the oxidation preventing agent and stabilizer, pellets are manufactured from the powder using a biaxial extruding mixer at a temperature between 200 and 230°C. The pellets are again extracted to measure properties

of the same.

Since the mixture of the monomers in the present invention changes the refraction coefficient in the resulting inventive resin, it is important to obtain a suitable monomer mixture to realize good transparency. That is, since a refractive coefficient of polybutadiene is approximately 1.518, an entire refractive coefficient of the grafted compound must be 1.510-1.526, preferably between 1.513 and 1.521. A refractive coefficient not reaching 1.510 or exceeding 1.527 is not suitable in the present invention.

The present invention is further explained in more detail with reference to the following examples. The invention can be utilized in various ways and is not intended to be confined to the examples.

#### [Example 1]

##### I. Manufacture of polybutadiene rubber latex

##### A) Manufacture of small aperture rubber latex

Added all at once to a nitrogen substituted polymerization autoclave were 110 parts by weight of ion exchange water; 100 parts by weight of 1,3-butadiene as a monomer; 1.2 parts by weight of rosin acid potassium salt as a emulsifying agent; 1.5 parts by weight of oleic acid potassium salt; 0.1 parts by weight of sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) as electrolyte; 0.5 parts by weight of potassium hydrogen carbonate ( $\text{KHCO}_3$ ); and 0.3 parts by weight of tertiary dodecyl mercaptan (TDDM) as a molecular weight controlling agent. Next, a reaction temperature was increased to 55°C, and after adding 0.3 parts by weight of persulfuric acid potassium as an initiator to begin the reaction, reaction

was performed for 10 hours. Following this step, 0.05 parts by weight of tertiary dodecyl mercaptan were again added to the mixture, after the same was reacted for 8 hours at 65°C. A rubber latex obtained using this method was analyzed using a method described below.

5 (1) Gel content and swelling index

After solidifying the rubber latex using diluted acid or metal salt, the rubber latex is washed then dried for 24 hours in a vacuum oven set at 60°C. Subsequently, the obtained rubber lumps are cut in small (approximately 1g) pieces using scissors, and the pieces are put into 100g of toluene then stored in  
10 a darkroom at room temperature for 48 hours. After storage, the material is separated into sol and gel, and a gel content and swelling index are measured using Equation 2 and Equation 3 below, respectively.

[Equation 2]

$$\text{Gel content(\%)} = \frac{\text{weight of insoluble portion (gel)}}{\text{weight of sample X}} \times 100$$

15

[Equation 3]

$$\text{Swelling index} = \frac{\text{weight of swelled gel}}{\text{weight of gel}}$$

(2) Particle diameter

Particle diameter is measured by a dynamic laser light scattering method  
20 using a Nicomp 370 HPL.

Using the above methods, the gel content of the particle diameter was 90%, the swelling index was 18, and the particle diameter was 1000 Å.

B) Manufacture of large aperture rubber latex (small aperture rubber

latex adhesion process)

Added to a reactor were 100 parts by weight of the small aperture rubber latex manufactured above. Next, at a stirring speed of 10 rpm and a temperature of 30°C, 3.5 parts by weight of an acetic acid aqueous solution were slowly  
5 added to the small aperture rubber latex over a period of one hour, after which stirring was discontinued and the mixture was left to stand for 30 minutes, thereby completing the manufacture of the large aperture rubber latex. The large aperture rubber latex obtained through an adhesion method in this manner had a particle diameter of 3000 Å, a gel content of 90%, and a swelling index of 17.

## 10 II. Manufacture of graft copolymerization material

Added all at once at a temperature of 50°C to a mixture of 8 parts by weight of the small aperture rubber latex and 10 parts by weight of the large aperture rubber latex were the components of Example 1 shown in Tables 1 and 2, i.e. 90 parts by weight of ion exchange water, 0.2 parts by weight of an oleic  
15 acid sodium emulsifying agent, 11.98 parts by weight of methylmethacrylate, 4.52 parts by weight of styrene, 4 parts by weight of acrylonitrile, 0.2 parts by weight of tertiary dodecyl mercaptan, 0.048 parts by weight of pyrophosphoric acid sodium, 0.012 parts by weight of dextrose, 0.001 parts by weight of primary iron sulfate, and 0.04 parts by weight of cumene hydroperoxide. The mixture  
20 was reacted while slowly increasing the temperature to 73°C over a period of 2 hours.

Next, continuously added to the above mixture over a period of 4 hours was an emulsifying agent mixture of 70 parts by weight of ion exchange water,

13

0.4 parts by weight of an oleic acid sodium emulsifying agent, 35.92 parts by weight of methylmethacrylate, 13.58 parts by weight of styrene, 12 parts by weight of acrylonitrile, 0.25 parts by weight of tertiary dodecyl mercaptan, 0.048 parts by weight of pyrophosphoric acid sodium, 0.012 parts by weight of dextrose, 0.001 parts by weight of primary iron sulfate, and 0.10 parts by weight of cumene hydroperoxide. The temperature was again increased to 76°C over a period of one hour to mature the mixture, thereby completing the reaction.

A polymerization conversion rate of the obtained latex was 99.8% and a solidification rate of the same was 0.12%. A powder was obtained by solidifying the latex using a calcium chloride aqueous solution then washing the same.

#### [Example 2]

The same method as in Example 1 was used to manufacture small and large aperture rubber latex, and graft copolymerization material, but a composition ratio for Example 2 shown in Tables 1 and 2 was used instead of that for Example 1.

#### [Comparative Examples 1-5]

The same method as in Example 1 was used to manufacture small and large aperture rubber latex, and graft copolymerization material, but a composition ratio for Comparative Examples 1-5 shown in Tables 1 and 2 was used instead of that for Example 1.

#### [Comparative Example 6]

The same method as in Example 1 was used to manufacture small aperture rubber latex and graft copolymerization material, but a composition ratio



for Comparative Example 6 shown in Tables 1 and 2 was used instead of that for Example 1. Large aperture rubber latex in Comparative Example 6 was manufactured as described below.

Manufacture of large aperture rubber latex (direct polymerization  
5 method)

Added all at once to a nitrogen substituted polymerization autoclave were 83 parts by weight of ion exchange water; 100 parts by weight of 1,3-butadiene; 1.2 parts by weight of rosin acid potassium salt as an emulsifying agent; 1.5 parts by weight of oleic acid potassium salt; 0.7 parts by weight of  
10 sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) as electrolyte; 0.8 parts by weight of potassium hydrogen carbonate ( $\text{KHCO}_3$ ); and 0.3 parts by weight of tertiary dodecyl mercaptan (TDDM) as a molecular weight controlling agent. Next, a reaction temperature was increased to  $65^\circ\text{C}$ , and after adding 0.3 parts by weight of persulfuric acid potassium as an initiator to initiate the reaction, reaction was  
15 performed for 45 hours while the temperature was raised to  $85^\circ\text{C}$ . A rubber latex obtained using this method was then analyzed, the results of which were a rubber latex having a particle diameter of  $3000\text{ \AA}$ , a gel content of 65%, and a swelling index of 31.

[Applied Example 1]

20 Added to 100 parts by weight of the graft copolymerization material of Example 1 were 0.1 parts by weight of an activator and 0.2 parts by weight of an oxidation preventing material. Next, using a biaxial extruding mixer, pellets were made from the graft copolymerization material and added components in a

cylinder oven set at 210°C. The pellets were extracted to make samples, and properties of the samples were measured, results of which are shown in Table 3.

[Applied Examples 2-8]

The same method as that used in Applied Example 1 was used, but instead of adding the materials to the graft copolymerization material of Example 1, the powder manufactured according to Example 2 and Comparative Examples 1-6 was used for Applied Examples 2-8, respectively. Results of analysis are shown in Table 3.

10 Table 1

1st Reaction	Example 1	Example 2	Comp. Example 1	Comp. Example 2	Comp. Example 3	Comp. Example 4	Comp. Example 5	Comp. Example 6
Ion Exch. Water	90	100	99	91	96	92	90	95
Small Apert. Rubber Latex	8	3	1	4	6	14	—	4
Large Apert. Rubber Latex	10	9	9	8	10	—	14	10 (direct method)
Oleic Acid Sodium	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Methylmethacrylate	11.98	—	—	15.8	—	—	14.38	19.2
Styrene	4.52	—	—	3.7	—	—	5.63	7.47
Acrylonitrile	4	—	—	2.5	—	—	2.5	2

Tertiary dodecyl mercaptan	0.2	—	—	0.2	—	—	0.2	0.2
Pyrophosphoric acid sodium	0.048	0.048	0.048	0.048	0.048	0.048	0.048	0.048
Dextrose	0.012	0.012	0.012	0.012	0.012	0.012	0.012	0.012
Primary iron sulfate	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Cumene hydroperoxide	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04
Add. Method	batch	batch	batch	batch	batch	batch	batch	batch
Reaction Time	2 hours	—	—	2 hours	—	—	2 hours	2 hours

## Table 2

[illegible]

n								
Pyrophosphoric acid sodium	0.048	0.048	0.048	0.048	0.048	0.048	0.048	0.048
Dextrose	0.012	0.012	0.012	0.012	0.012	0.012	0.012	0.012
Primary iron sulfate	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Cumene hydroperoxide	0.1	0.11	0.1	0.1	0.1	0.1	0.1	0.1
Add. Method	Continuous	continuous	continuous	continuous	continuous	continuous	continuous	continuous
Reaction Time	4 hours	6 hours	6 hours	4 hours	6 hours	6 hours	4 hours	4 hours
Polymer. Conv. Rate(%)	99.8	99.0	99.7	99.7	99.5	99.5	99.3	99.8
Solidification (%)	0.12	0.10	0.10	0.10	0.15	0.10	0.10	0.10

Table 3

	Applied Ex. 1	Applied Ex. 2	Applied Ex. 3	Applied Ex. 4	Applied Ex. 5	Applied Ex. 6	Applied Ex. 7	Applied Ex. 8
Graft copolymerization material	Example 1	Example 2	Comp. Example 1	Comp. Example 2	Comp. Example 3	Comp. Example 4	Comp. Example 5	Comp. Example 6
Notch Isod Shock Strength ASTM D-256	16	11	7	10	14	3	13	13
Fluidity Index ASTM D-1238	14	22	25	22	16	16	18	19

18

Haze value ASTM D-1003	3.5	3	3.9	20	24	2.4	8.8	11.2
Chemical Resist. (ethanol solution) (after 14-day storage)	Extremely High	High	Low	High	High	High	High	High

Although preferred examples of the present invention have been described in detail hereinabove, it should be clearly understood that many variations and/or modifications of the basic inventive concepts herein taught  
5 which may appear to those skilled in the present art will still fall within the spirit and scope of the present invention, as defined in the appended claims.

**WHAT IS CLAIMED IS:**

1. A thermoplastic transparent resin composition comprising:  
3-15 parts by weight of small aperture polybutadiene rubber latex;  
5-25 parts by weight of large aperture polybutadiene rubber latex;  
5 40-70 parts by weight of a methacrylic acid alkylester compound or an  
acrylic acid alkylester compound;  
15-30 parts by weight of an aromatic vinyl compound; and  
1-20 parts by weight of a vinylcian compound.
2. The resin composition of claim 1 wherein the composition further  
10 comprises 0.2-0.6 parts by weight of an emulsifying agent, 0.2-0.6 parts by  
weight of a molecular weight controlling agent, and 0.05-0.3 parts by weight of a  
polymerization starter.
3. The resin composition of claim 1 wherein the small aperture  
polybutadiene rubber latex has a particle diameter of 600-1500 Å, a gel content  
15 of 70-95%, and a swelling index of 12-30.
4. The resin composition of claim 1 wherein the large aperture  
polybutadiene rubber latex has a particle diameter of 2600-5000 Å, a gel content  
of 70-95%, and a swelling index of 12-30,
5. The resin composition of claim 1 wherein the methacrylic acid  
20 alkylester compound and the acrylic acid alkylester compound are  
methylmethacrylate.
6. The resin composition of claim 1 wherein the aromatic vinyl compound  
is selected from the group consisting of styrene,  $\alpha$ -methylstyrene, o-

ethylstyrene, p-ethylstyrene, and vinyl toluene.

7. The resin composition of claim 1 wherein the vinylcian compound is selected from the group consisting of acrylonitrile, methacrylonitrile, and ethacrylonitrile.

5 8. A method of manufacturing a thermoplastic transparent resin composition comprising the steps of:

a) producing a small aperture polybutadiene rubber latex having an average particle diameter of 600-1500  $\overset{\circ}{\text{\AA}}$ , a gel content of 70-95%, and a swelling index of 12-30 by reacting butadiene at 55-70°C using a polymerization  
10 initiator;

b) producing a large aperture polybutadiene rubber latex having a particle diameter of 2600-5000  $\overset{\circ}{\text{\AA}}$ , a gel content of 70-95%, and a swelling index of 12-30 by enlarging the small aperture polybutadiene rubber latex particles; and

15 c) performing graft copolymerization at 65-80°C by continuously or separately adding 3-15 parts by weight of the small aperture polybutadiene rubber latex of step a), 5-25 parts by weight of the large aperture polybutadiene rubber latex of step b), 40-70 parts by weight of a methacrylic acid alkylester compound or an acrylic acid alkylester compound, 15-30 parts by weight of an  
20 aromatic vinyl compound, and 1-20 parts by weight of a vinylcian compound.

9. The method of claim 8 wherein the methacrylic acid alkylester compound and the acrylic acid alkylester compound are methylmethacrylate.

10. The method of claim 8 wherein the aromatic vinyl compound is

21

selected from the group consisting of styrene,  $\alpha$ -methylstyrene, o-ethylstyrene, p-ethylstyrene, and vinyl toluene.

11. The method of claim 8 wherein the vinylcyan compound is selected from the group consisting of acrylonitrile, methacrylonitrile, and ethacrylonitrile.

5 12. The method of claim 8 wherein in step c), a total refraction coefficient of the compound, excluding the polybutadiene rubber latex, is between 1.510 and 1.526.



## PATENT COOPERATION TREATY

PCT

From the INTERNATIONAL BUREAU

NOTIFICATION CONCERNING  
SUBMISSION OR TRANSMITTAL  
OF PRIORITY DOCUMENT

(PCT Administrative Instructions, Section 411)

To:

KIM, Seong-Ki  
9th Teheran Boulevard  
825-33, Yoksam-dong  
Kangnam-gu  
Seoul 135-080  
RÉPUBLIQUE DE CORÉE

Date of mailing (day/month/year) 09 April 1999 (09.04.99)	<b>IMPORTANT NOTIFICATION</b>
Applicant's or agent's file reference OPP990070KR	
International application No. PCT/KR99/00101	International filing date (day/month/year) 04 March 1999 (04.03.99)
International publication date (day/month/year) Not yet published	Priority date (day/month/year) 29 October 1998 (29.10.98)
Applicant LG CHEMICAL LTD. et al	

1. The applicant is hereby notified of the date of receipt (except where the letters "NR" appear in the right-hand column) by the International Bureau of the priority document(s) relating to the earlier application(s) indicated below. Unless otherwise indicated by an asterisk appearing next to a date of receipt, or by the letters "NR", in the right-hand column, the priority document concerned was submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b).
2. This updates and replaces any previously issued notification concerning submission or transmittal of priority documents.
3. An asterisk(\*) appearing next to a date of receipt, in the right-hand column, denotes a priority document submitted or transmitted to the International Bureau but not in compliance with Rule 17.1(a) or (b). In such a case, **the attention of the applicant is directed** to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.
4. The letters "NR" appearing in the right-hand column denote a priority document which was not received by the International Bureau or which the applicant did not request the receiving Office to prepare and transmit to the International Bureau, as provided by Rule 17.1(a) or (b), respectively. In such a case, **the attention of the applicant is directed** to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

<u>Priority date</u>	<u>Priority application No.</u>	<u>Country or regional Office or PCT receiving Office</u>	<u>Date of receipt of priority document</u>
29 Octo 1998 (29.10.98)	1998/45699	KR	19 Marc 1999 (19.03.99)

The International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland

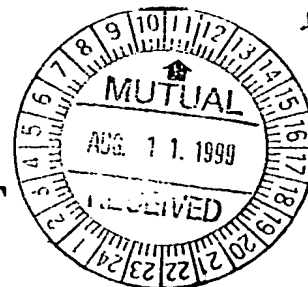
Facsimile No. (41-22) 740.14.35

Authorized officer

S. Cruz

Telephone No. (41-22) 338.83.38

# PATENT COOPERATION TREATY



From the INTERNATIONAL SEARCHING AUTHORITY

## PCT

### NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT OR THE DECLARATION

(PCT Rule 44.1)

To:

KIM, Seong-ki  
9th Floor, Teheran Bd.,  
825-33, Yoksam-dong,  
Kangnam-ku  
Seoul 135-080  
Republic of Korea

Date of mailing  
(day/month/year) 5 Aug. 1999 (05.08.99)

Applicant's or agent's file reference  
OPP990070KR

**FOR FURTHER ACTION** See paragraphs 1 and 4 below

International application No.  
PCT / KR 99/00101

International filing date  
(day/month/year) 4 Mrz. 1999 (04.03.99)

Applicant

LG CHEMICAL LTD. et al.

1. ☒ The applicant is hereby notified that the international search report has been established and is transmitted herewith.

**Filing of amendments and statement under Article 19:**

The applicant is entitled, if he so wishes, to amend the claims of the international application (see Rule 46):

**When?** The time limit for filing such amendments is normally two months from the date of transmittal of the international search report; however, for more details, see the notes on the accompanying sheet.

**Where?** Directly to the International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland  
Facsimile No.: (41-22) 740.14.35

For more detailed instructions, see the notes on the accompanying sheet.

2. ☐ The applicant is hereby notified that no international search report will be established and that the declaration under Article 17(2)(a) to that effect is transmitted herewith.

3. ☐ With regard to the protest against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:
- ☐ the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.
  - ☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Further action(s):** The applicant is reminded of the following:

Shortly after 18 months from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90bis.1 and 90bis.3, respectively, before the completion of the technical preparations for international publication.

Within 19 months from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later).

Within 20 months from the priority date, the applicant must perform the prescribed acts for entry into the national phase before all designated Offices which have not been elected in the demand or in a later election within 19 months from the priority date or could not be elected because they are not bound by Chapter II.

Name and mailing address of the ISA/  
AT  
AUSTRIAN PATENT OFFICE  
Kohlmarkt 8-10  
A-1014 Vienna  
Facsimile No. +43 / 1 / 534 24 - 200

Authorized officer

Koch

Telephone No. +43 / 1 / 534 24 - 450

## NOTES TO FORM PCT/ISA/220

These Notes are intended to give the basic instructions concerning the filing of amendments under Article 19. The Notes are based on the requirements of the Patent Cooperation Treaty, the Regulations and the Administrative Instructions under that Treaty. In case of discrepancy between these Notes and those requirements, the latter are applicable. For more detailed information, see also the *PCT Applicant's Guide*, a publication of WIPO.

In these Notes, "Article," "Rule" and "Section" refer to the provisions of the PCT, the PCT Regulations and the PCT Administrative Instructions, respectively.

### INSTRUCTIONS CONCERNING AMENDMENTS UNDER ARTICLE 19

The applicant has, after having received the international search report, one opportunity to amend the claims of the international application. It should however be emphasized that, since all parts of the international application (claims, description and drawings) may be amended during the international preliminary examination procedure, there is usually no need to file amendments of the claims under Article 19 except where, e.g. the applicant wants the latter to be published for the purposes of provisional protection or has another reason for amending the claims before international publication. Furthermore, it should be emphasized that provisional protection is available in some States only.

#### What parts of the international application may be amended ?

Under Article 19, only the claims may be amended.

During the international phase, the claims may also be amended (or further amended) under Article 34 before the International Preliminary Examining Authority. The description and drawings may only be amended under Article 34 before the International Preliminary Examining Authority.

Upon entry into the national phase, all parts of the international application may be amended under Article 28 or, where applicable, Article 41.

**When ?** Within 2 months from the date of transmittal of the international search report or 16 months from the priority date, whichever time limit expires later. It should be noted, however, that the amendments will be considered as having been received on time if they are received by the International Bureau after the expiration of the applicable time limit but before the completion of the technical preparations for international publication (Rule 46.1).

#### Where not to file the amendments ?

The amendments may only be filed with the International Bureau and not with the receiving Office or the International Searching Authority (Rule 46.2).

Where a demand for international preliminary examination has been/is filed, see below.

**How ?** Either by cancelling one or more entire claims, by adding one or more new claims or by amending the text of one or more of the claims as filed.

A replacement sheet must be submitted for each sheet of the claims which, on account of an amendment or amendments, differs from the sheet originally filed.

All the claims appearing on a replacement sheet must be numbered in Arabic numerals. Where a claim is cancelled, no renumbering of the other claims is required. In all cases where claims are renumbered, they must be renumbered consecutively (Administrative Instructions, Section 205(b)).

The amendments must be made in the language in which the international application is to be published.

#### What documents must/may accompany the amendments ?

Letter (Section 205(b)):

The amendments must be submitted with a letter.

The letter will not be published with the international application and the amended claims. It should not be confused with the "Statement under Article 19(1)" (see below, under "Statement under Article 19(1)").

The letter must be in English or French, at the choice of the applicant. However, if the language of the international application is English, the letter must be in English; if the language of the international application is French, the letter must be in French.

## NOTES TO FORM PCT/ISA/220 (continued)

The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped), whether

- (i) the claim is unchanged;
- (ii) the claim is cancelled;
- (iii) the claim is new;
- (iv) the claim replaces one or more claims as filed;
- (v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

1. [Where originally there were 48 claims and after amendment of some claims there are 51]:  
"Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
2. [Where originally there were 15 claims and after amendment of all claims there are 11]:  
"Claims 1 to 15 replaced by amended claims 1 to 11."
3. [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:  
"Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or  
"Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
4. [Where various kinds of amendments are made]:  
"Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

### "Statement under Article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

**It must be in the language in which the international application is to be published.**

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

### Consequence if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments and any accompanying statement, under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the time of filing the amendments (and any statement) with the International Bureau, also file with the International Preliminary Examining Authority a copy of such amendments (and of any statement) and, where required, a translation of such amendments for the procedure before that Authority (see Rules 55.3(a) and 62.2, first sentence). For further information, see the Notes to the demand form (PCT/IPEA/401).

### Consequence with regard to translation of the international application for entry into the national phase

The applicant's attention is drawn to the fact that, upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see the *PCT Applicant's Guide*, Volume II.

## PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>OPP 990070 KR</b>	FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/KR 99/00101</b>	International filing date (day/month/year) <b>04 March 1999 (04.03.99)</b>	(Earliest) Priority Date (day/month/year) <b>29 October 1998 (29.10.98)</b>
Applicant <b>LG CHEMICAL LTD.</b>		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

## 1. Basis of the report

a. With regard to the language, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international search was carried out on the basis of the sequence listing:

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☐ Unity of invention is lacking (See Box II).

4. With regard to the title,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the abstract,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the drawings to be published with the abstract is Figure No.: \_\_\_\_\_

☐ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☐ None of the figures.

# PCT

## REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

For receiving Office use only

International Application No.

International Filing Date

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference  
(if desired) (12 characters maximum)

OPP990070KR

Box No. I TITLE OF INVENTION THERMOPLASTIC TRANSPARENT RESIN COMPOSITION  
AND METHOD OF MANUFACTURING SAME

Box No. II APPLICANT

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

LG CHEMICAL LTD.

LG Twin Towers, 20 Yoido-dong, Yongsungpo-gu,  
Seoul 150-721, Republic of Korea

☐ This person is also inventor.

Telephone No.

042-866-2072

Facsimile No.

042-863-2053

Teleprinter No.

State (that is, country) of nationality:

KR

State (that is, country) of residence:

KR

This person is applicant  
for the purposes of:

☐ all designated  
States

☒ all designated States except  
the United States of America

☐ the United States  
of America only

☐ the States indicated in  
the Supplemental Box

Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

YOO, Keon-Hoon

Hanbit Apt. 123-801, Eoeun-dong,

Yuseong-gu, Daejeon-city 305-333

Republic of Korea

This person is:

☐ applicant only

☒ applicant and inventor

☐ inventor only (If this check-box  
is marked, do not fill in below.)

State (that is, country) of nationality:

KR

State (that is, country) of residence:

KR

This person is applicant  
for the purposes of:

☐ all designated  
States

☐ all designated States except  
the United States of America

☒ the United States  
of America only

☐ the States indicated in  
the Supplemental Box

☒ Further applicants and/or (further) inventors are indicated on a continuation sheet.

Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The person identified below is hereby/has been appointed to act on behalf  
of the applicant(s) before the competent International Authorities as:

☒ agent

☐ common representative

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

KIM, Seong-Ki

9th Teheran Bd., 825-33

Yoksam-dong, Kangnam-gu

Seoul 135-080, Republic of Korea

Telephone No.

02-3458-0800

Facsimile No.

02-539-0758

Teleprinter No.

☐ Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

Continuation of Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)	
<i>If none of the following sub-boxes is used, this sheet should not be included in the request.</i>	
<p>Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)</p> <p>SHIN, Yang-Hyun 29, Ansan-dong, Yochon-city, Chollanam-do 555-050 Republic of Korea</p>	<p>This person is:</p> <p><input type="checkbox"/> applicant only</p> <p><input checked="" type="checkbox"/> applicant and inventor</p> <p><input type="checkbox"/> inventor only (If this check-box is marked, do not fill in below.)</p>
State (that is, country) of nationality: KR	State (that is, country) of residence: KR
<p>This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input checked="" type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box</p>	
<p>Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)</p> <p>LEE, Chan-Hong Ruckyhana Apt. 102-901, Shinseong-dong, Yuseong-gu, Daejeon-city 305-345 Republic of Korea</p>	<p>This person is:</p> <p><input type="checkbox"/> applicant only</p> <p><input checked="" type="checkbox"/> applicant and inventor</p> <p><input type="checkbox"/> inventor only (If this check-box is marked, do not fill in below.)</p>
State (that is, country) of nationality: KR	State (that is, country) of residence: KR
<p>This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input checked="" type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box</p>	
<p>Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)</p>	<p>This person is:</p> <p><input type="checkbox"/> applicant only</p> <p><input type="checkbox"/> applicant and inventor</p> <p><input type="checkbox"/> inventor only (If this check-box is marked, do not fill in below.)</p>
State (that is, country) of nationality:	State (that is, country) of residence:
<p>This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box</p>	
<p>Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)</p>	<p>This person is:</p> <p><input type="checkbox"/> applicant only</p> <p><input type="checkbox"/> applicant and inventor</p> <p><input type="checkbox"/> inventor only (If this check-box is marked, do not fill in below.)</p>
State (that is, country) of nationality:	State (that is, country) of residence:
<p>This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box</p>	
<p><input type="checkbox"/> Further applicants and/or (further) inventors are indicated on another continuation sheet.</p>	

## Box No.V DESIGNATION OF STATES

The following designations are hereby made under Rule 4.9(a) (mark the applicable check-boxes; at least one must be marked):

## Regional Patent

- ☐ AP ARIPO Patent: GH Ghana, GM Gambia, KE Kenya, LS Lesotho, MW Malawi, SD Sudan, SZ Swaziland, UG Uganda, ZW Zimbabwe, and any other State which is a Contracting State of the Harare Protocol and of the PCT
- ☐ EA Eurasian Patent: AM Armenia, AZ Azerbaijan, BY Belarus, KG Kyrgyzstan, KZ Kazakhstan, MD Republic of Moldova, RU Russian Federation, TJ Tajikistan, TM Turkmenistan, and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT
- ☒ EP European Patent: AT Austria, BE Belgium, CH and LI Switzerland and Liechtenstein, CY Cyprus, DE Germany, DK Denmark, ES Spain, FI Finland, FR France, GB United Kingdom, GR Greece, IE Ireland, IT Italy, LU Luxembourg, MC Monaco, NL Netherlands, PT Portugal, SE Sweden, and any other State which is a Contracting State of the European Patent Convention and of the PCT
- ☐ OA OAPI Patent: BF Burkina Faso, BJ Benin, CF Central African Republic, CG Congo, CI Côte d'Ivoire, CM Cameroon, GA Gabon, GN Guinea, GW Guinea-Bissau, ML Mali, MR Mauritania, NE Niger, SN Senegal, TD Chad, TG Togo, and any other State which is a member State of OAPI and a Contracting State of the PCT (if other kind of protection or treatment desired, specify on dotted line)

## National Patent (if other kind of protection or treatment desired, specify on dotted line):


- |   |   |
|---|---|
| <input type="checkbox"/> AL Albania                               | <input type="checkbox"/> LS Lesotho                                   |
| <input type="checkbox"/> AM Armenia                               | <input type="checkbox"/> LT Lithuania                                 |
| <input type="checkbox"/> AT Austria                               | <input type="checkbox"/> LU Luxembourg                                |
| <input type="checkbox"/> AU Australia                             | <input type="checkbox"/> LV Latvia                                    |
| <input type="checkbox"/> AZ Azerbaijan                            | <input type="checkbox"/> MD Republic of Moldova                       |
| <input type="checkbox"/> BA Bosnia and Herzegovina                | <input type="checkbox"/> MG Madagascar                                |
| <input type="checkbox"/> BE Barbados                              | <input type="checkbox"/> MK The former Yugoslav Republic of Macedonia |
| <input type="checkbox"/> BG Bulgaria                              | <input type="checkbox"/> MN Mongolia                                  |
| <input type="checkbox"/> BR Brazil                                | <input type="checkbox"/> MW Malawi                                    |
| <input type="checkbox"/> BY Belarus                               | <input type="checkbox"/> MX Mexico                                    |
| <input type="checkbox"/> CA Canada                                | <input type="checkbox"/> NO Norway                                    |
| <input type="checkbox"/> CH and LI Switzerland and Liechtenstein  | <input type="checkbox"/> NZ New Zealand                               |
| <input type="checkbox"/> CN China                                 | <input type="checkbox"/> PL Poland                                    |
| <input type="checkbox"/> CU Cuba                                  | <input type="checkbox"/> PT Portugal                                  |
| <input type="checkbox"/> CZ Czech Republic                        | <input type="checkbox"/> RO Romania                                   |
| <input type="checkbox"/> DE Germany                               | <input type="checkbox"/> RU Russian Federation                        |
| <input type="checkbox"/> DK Denmark                               | <input type="checkbox"/> SD Sudan                                     |
| <input type="checkbox"/> EE Estonia                               | <input type="checkbox"/> SE Sweden                                    |
| <input type="checkbox"/> ES Spain                                 | <input type="checkbox"/> SG Singapore                                 |
| <input type="checkbox"/> FI Finland                               | <input type="checkbox"/> SI Slovenia                                  |
| <input type="checkbox"/> GB United Kingdom                        | <input type="checkbox"/> SK Slovakia                                  |
| <input type="checkbox"/> GD Grenada                               | <input type="checkbox"/> SL Sierra Leone                              |
| <input type="checkbox"/> GE Georgia                               | <input type="checkbox"/> TJ Tajikistan                                |
| <input type="checkbox"/> GH Ghana                                 | <input type="checkbox"/> TM Turkmenistan                              |
| <input type="checkbox"/> GM Gambia                                | <input type="checkbox"/> TR Turkey                                    |
| <input type="checkbox"/> HR Croatia                               | <input type="checkbox"/> TT Trinidad and Tobago                       |
| <input type="checkbox"/> HU Hungary                               | <input type="checkbox"/> UA Ukraine                                   |
| <input type="checkbox"/> ID Indonesia                             | <input type="checkbox"/> UG Uganda                                    |
| <input type="checkbox"/> IL Israel                                | <input checked="" type="checkbox"/> US United States of America       |
| <input type="checkbox"/> IN India                                 | <input type="checkbox"/> UZ Uzbekistan                                |
| <input type="checkbox"/> IS Iceland                               | <input type="checkbox"/> VN Viet Nam                                  |
| <input type="checkbox"/> JP Japan                                 | <input type="checkbox"/> YU Yugoslavia                                |
| <input type="checkbox"/> KE Kenya                                 | <input type="checkbox"/> ZW Zimbabwe                                  |
| <input type="checkbox"/> KG Kyrgyzstan                            |   |
| <input type="checkbox"/> KP Democratic People's Republic of Korea |   |
| <input type="checkbox"/> KR Republic of Korea                     |   |
| <input type="checkbox"/> KZ Kazakhstan                            |   |
| <input type="checkbox"/> LC Saint Lucia                           |   |
| <input type="checkbox"/> LK Sri Lanka                             |   |
| <input type="checkbox"/> LR Liberia                               |   |

Check-boxes reserved for designating States (for the purposes of a national patent) which have become party to the PCT after issuance of this sheet:

- ☐ .....
- ☐ .....
- ☐ .....

**Precautionary Designation Statement:** In addition to the designations made above, the applicant also makes under Rule 4.9(b) all other designations which would be permitted under the PCT except any designation(s) indicated in the Supplemental Box as being excluded from the scope of this statement. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit. (Confirmation of a designation consists of the filing of a notice specifying that designation and the payment of the designation and confirmation fees. Confirmation must reach the receiving Office within the 15-month time limit.)



<b>Box No. VI PRIORITY CLAIM</b>		<input type="checkbox"/> Further priority claims are indicated in the Supplemental Box.		
Filing date of earlier application (day/month/year)	Number of earlier application	Where earlier application is:		
		national application: country	regional application: regional Office	international application: receiving Office
item (1) 29/October/1998 (29. 10. 1998)	98-45699	KR		
item (2)				
item (3)				
<input type="checkbox"/> The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) (only if the earlier application was filed with the Office which for the purposes of the present international application is the receiving Office) identified above as item(s):				
<i>* Where the earlier application is an ARIPO application, it is mandatory to indicate in the Supplemental Box at least one country party to the Paris Convention for the Protection of Industrial Property for which that earlier application was filed (Rule 4.10(b)(ii)). See Supplemental Box.</i>				
<b>Box No. VII INTERNATIONAL SEARCHING AUTHORITY</b>				
Choice of International Searching Authority (ISA) (if two or more International Searching Authorities are competent to carry out the international search, indicate the Authority chosen; the two-letter code may be used):		Request to use results of earlier search; reference to that search (if an earlier search has been carried out by or requested from the International Searching Authority):		
ISA /		Date (day/month/year)	Number	Country (or regional Office)
<b>Box No. VIII CHECK LIST; LANGUAGE OF FILING</b>				
This international application contains the following number of sheets: request : 4 description (excluding sequence listing part) : 18 claims : 3 abstract : 1 drawings : sequence listing part of description : Total number of sheets : 26		This international application is accompanied by the item(s) marked below: 1. <input checked="" type="checkbox"/> fee calculation sheet 2. <input checked="" type="checkbox"/> separate signed power of attorney 3. <input type="checkbox"/> copy of general power of attorney; reference number, if any: 4. <input type="checkbox"/> statement explaining lack of signature 5. <input checked="" type="checkbox"/> priority document(s) identified in Box No. VI as item(s): (1) 6. <input type="checkbox"/> translation of international application into (language): 7. <input type="checkbox"/> separate indications concerning deposited microorganism or other biological material 8. <input type="checkbox"/> nucleotide and/or amino acid sequence listing in computer readable form 9. <input type="checkbox"/> other (specify):		
Figure of the drawings which should accompany the abstract:		Language of filing of the international application: English		
<b>Box No. IX SIGNATURE OF APPLICANT OR AGENT</b>				
Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request).				
KIM, Seong-Ki 				

For receiving Office use only	
1. Date of actual receipt of the purported international application: 3. Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application: 4. Date of timely receipt of the required corrections under PCT Article 11(2): 5. International Searching Authority (if two or more are competent): ISA /	2. Drawings: <input type="checkbox"/> received: <input type="checkbox"/> not received: 6. <input type="checkbox"/> Transmittal of search copy delayed until search fee is paid.

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Date of receipt of _____ copy by the International _____

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/KR 99/00101

## A. CLASSIFICATION OF SUBJECT MATTER

IPC<sup>7</sup>: C 08 L 33/06; (C 08 L 33/06; C 08 L 25:08; C 08 L 9:00; C 08 L 33:20)

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC<sup>7</sup>: C 08 L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

Questel Telesystems, London, File: WPI

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4 242 469 A (SCHMITT et al.), 30 December 1980 (30.12.80), examples; claims.	1-12
A	US 4 228 256 A (SCHMITT), 14 October 1980 (14.10.80), examples; claims.	1-12
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A	Derwent Accession No 81-41015 D[23], Questel Telesystems (WPI), Derwent Publications Ltd. London, abstract; & JP 56-041 216 A (JAPAN SYNTHETIC RUBBER CO., LTD.), DW 8123 (17.04.81)	1-12

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

\* Special categories of cited documents:

„A“ document defining the general state of the art which is not considered to be of particular relevance

„E“ earlier application or patent but published on or after the international filing date

„L“ document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

„O“ document referring to an oral disclosure, use, exhibition or other means

„P“ document published prior to the international filing date but later than the priority date claimed

„T“ later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

„X“ document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

„Y“ document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

„&“ document member of the same patent family

Date of the actual completion of the international search

27 July 1999 (27.07.99)

Date of mailing of the international search report

05 August 1999 (05.08.99)

Name and mailing address of the ISA/AT

Austrian Patent Office  
Kohlmarkt 8-10; A-1014 Vienna  
Facsimile No. 1/53424/200

Authorized officer

Tengler

Telephone No. 1/53424/213

INTERNATIONAL SEARCH REPORT  
Inform: on patent family members

International application No.  
PCT/KR 99/00101

In Recherchebericht angeführtes Patentedokument Patent document cited in search report Document de brevet cité dans le rapport de recherche	Datum der Veröffentlichung Publication date Date de publication	Mitglied(er) der Patentfamilie Patent family member(s) Membre(s) de la famille de brevets	Datum der Veröffentlichung Publication date Date de publication
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